Spin-glass behavior in CeCu2-type uranium compound U2AuGa3

doi: 10.1063/1.2081130

Spin-glass behavior in $CeCu_2$-type uranium compound U_2AuGa_3

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(Received 3 May 2005; accepted 11 August 2005; published online 27 September 2005)

We present the results of ac and dc susceptibility, magnetization, magnetic relaxation, specific heat, and electrical resistivity measurements on U_2AuGa_3, an orthorhombic $CeCu_2$-type nonmagnetic atom disorder system. These data clearly indicate that U_2AuGa_3 undergoes a spin glass phase transition at a static freezing temperature T_s=23.6 K, in spite of the lack of triangular magnetic structure. It is observed that the variation of the characteristic temperature T_{ir} (the bifurcation point between field-cooled and zero-field-cooled susceptibilities) with applied field H for U_2AuGa_3 is not consistent with the "AT line," but follows a $T_{ir} \propto -H^{2/5}$ law. The observed spin glass behavior and the formation of frustrated magnetic interactions in U_2AuGa_3 are discussed in a magnetic cluster model. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2081130]

Spin glass (SG) behavior observed in uranium-based nonmagnetic atom disorder (NMAD) compounds $U_2T\,Si_3$ (T is a d-electron transition metal)[1] have attracted much interest in recent years. These compounds (expect U_2CuSi_3) crystallize in the hexagonal AlB_2-type structure, where T and Si atoms are randomly distributed into the trigonal prisms of a primitive hexagonal array of uranium atoms.[2] Frustration of the magnetic interactions, *one necessary condition for SG state*, could be induced by this topology in the case of antiferromagnetic (AF) coupling between nearest neighbours. Such an origin of SG behavior in $U_2T\,Si_3$ is evidently different from that in amorphous or diluted metallic spin glass and relates to the inherent nature of the AlB_2-type crystal structure. Recently, we have been paying special attention to U_2TGa_3, another series of NMAD uranium compounds with the $CeCu_2$-type structure *without a triangular magnetic lattice* because of their characteristic magnetic and structural features.[3,4] In particular, we have confirmed the existence of another member, namely U_2AuGa_3, in the U_2TGa_3 family and performed a detailed investigation on its magnetic properties. In this letter, we report the discovery of SG behavior in the U_2AuGa_3 sample and present the dynamical parameters characterizing the frozen state of this system. The possible mechanism of frustrated magnetic interactions in $CeCu_2$-type U_2AuGa_3 lacking a triangular magnetic structure is discussed in a magnetic cluster model.

The polycrystalline sample of U_2AuGa_3 was synthesized by arc melting the constituent elements with high purities (3N for U, 4N for Au, and 6N for Ga) in a purified argon atmosphere, then sealing in an evacuated quartz glass tube, and annealing at 800 ° C for a week. X-ray diffraction measurements show the pattern structure of U_2AuGa_3 as the same as that observed for our U_2PdGa_3 and U_2PtGa_3 samples, and the diffraction lines can be indexed based on the disordered orthorhombic $CeCu_2$-type structure model (space group *Imma*) with U atoms on the 4*e* sites and Au and Ga atoms statistically distributed over the 8*h* sites. Note that for U_2PdGa_3 and U_2PtGa_3, the $CeCu_2$-type crystal structure has also been confirmed by neutron-powder-diffraction (NPD) measurements.[4] The determined room-temperature lattice constants of U_2AuGa_3 are a=4.435 Å, b=7.079 Å, and c=7.799 Å. The ac susceptibility, low field dc magnetization, and magnetic relaxation were measured using a SQUID magnetometer. High-field magnetization experiment up to 115 kOe was carried out at 5 K using a vibrating sample magnetometer. The adiabatic heat pulse method and standard four-terminal method were employed for specific heat and electrical resistivity measurements, respectively.

Figure 1 shows the temperature dependence of the in-phase component $\chi'_{ac}(T,\omega)$ of the ac susceptibility of U_2AuGa_3 between 15 and 34 K at the frequency range $0.1 \leqslant \omega/2\pi \leqslant 1000$ Hz. The χ'_{ac} curve exhibits a characteristic pronounced maximum with amplitude and position $[T_f(\omega)]$ depending on the frequency of the ac magnetic field. As ω increases, T_f increases from 24.4 K at $\omega/2\pi$=0.1 Hz to 25.8 K at $\omega/2\pi$=1000 Hz. This is a typical behavior characteristic of SG materials suggesting the formation of the SG state in U_2AuGa_3. The initial frequency shift of T_f defined as $\delta T_f = \Delta T_f/(T_f \Delta \log \omega)$ is determined to be δT_f=0.01 comparable to the typical values (from a few thousandths to a few

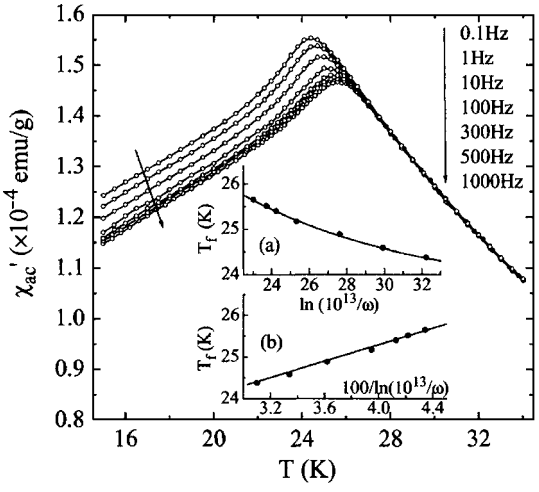

FIG. 1. Temperature dependence of the real component (χ'_{ac}) of ac susceptibility for U_2AuGa_3 measured at different frequencies. The frequency dependencies of the dynamic spin freezing temperature T_f plotted as T_f vs $\ln(10^{13}/\omega)$ and T_f vs $100/\ln(10^{13}/\omega)$ are shown in the inset (a) and the inset (b), respectively.

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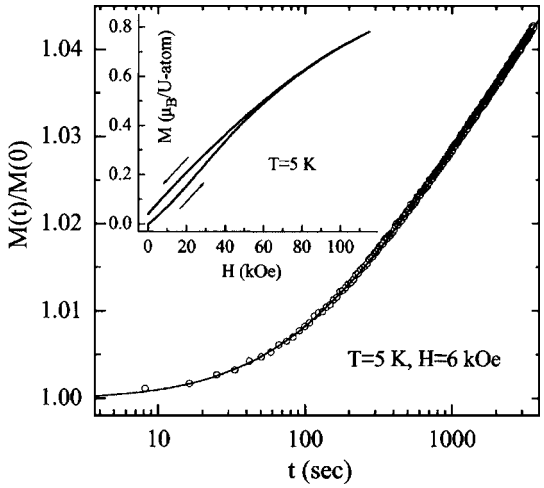

FIG. 2. Time dependence of magnetization of U_2AuGa_3, plotted as $M(t)/M(0)$ vs t, measured at 5 K in magnetic field of 6.0 kOe. The solid line represents least squares fit using equation $M_{\mathrm{IRM}}(t)=M_0+S\ln(t+t_0)$. The inset displays the $M(H)$ data up to 115 kOe measured at 5 K.

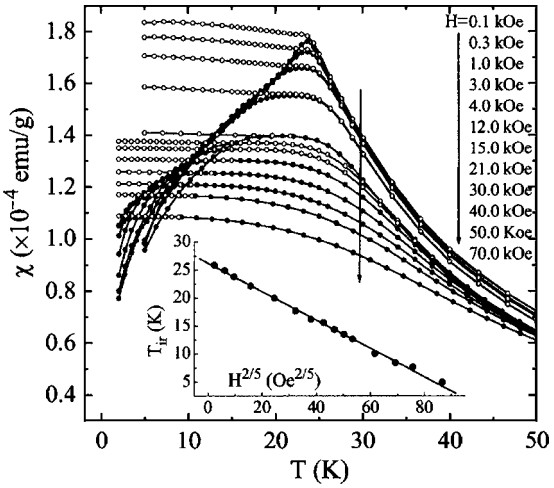

FIG. 3. Comparison of the low-temperature FC (open symbols) and ZFC (filled symbols) susceptibilities ($\chi=M/H$) for U_2AuGa_3 measured in various magnetic fields. The inset shows the field dependence of the characteristic temperature T_{ir} (from where magnetic irreversibility occurs), plotted as T_{ir} vs $H^{2/5}$.

hundredths) for most spin glasses.[5] In contrast, we do not find any upward shift of the peak position T_N in the χ'_{ac} curves for long-range AF ordered compounds U_2PdGa_3 and U_2PtGa_3 up to 1000 Hz (not shown here). In order to estimate the dynamical parameters characterizing the SG state of U_2AuGa_3, the obtained $T_f(\omega)$ data are fitted to the standard expression $\tau_{\max}=\tau_0[(T_f-T_s)/T_s]^{-z\nu}$ (the critical slowing down)[6] and to the Vogel-Fulcher Law[7,8] $\omega=\omega_0 \exp[-E_a/k_B(T_f-T_0)]$, respectively. Following Tholence,[9] $\tau_0=1/\omega_0=10^{-13}$ s was kept fixed, the fit using the former equation [solid line in the inset (a) of Fig. 1] yields the static freezing temperature $T_s=23.6$ K and the critical (dynamical) exponent $z\nu=9.4$, and the fit using the latter one [solid line in the inset (b) of Fig. 1] yields the activation energy $E_a\approx 4.4\ k_BT_s$ and the Vogel-Fulcher temperature $T_0=21.2$ K. It is worth notice that many experimental results give $z\nu=4$–12 for different SG systems, while $z\nu$ is usually around 2 for conventional phase transitions.[5]

Remanence and long-time magnetic relaxation effects are also the characteristic features of spin glasses. The inset of Fig. 2 shows the high-field magnetization $M(H)$ of U_2AuGa_3 at 5 K up to 115 kOe. Hysteresis effect is clearly observed, while a remanent magnetization of about 0.04 μ_B/U is detected at zero field. From a careful measurement of the hysteresis loop using a SQUID magnetometer, we determine the coercive field H_C of U_2AuGa_3 to be 4.5 kOe at 5 K. We investigated the magnetic relaxation behavior $[M(t)]$ of U_2AuGa_3 at 5 K in a field of 6.0 kOe ($\geqslant H_C=4.5$ kOe). The sample was first cooled in zero field from 150 K to 5 K, then a field of 6.0 kOe was applied, and the recording started immediately just as the field stabilized ($t=0$). As seen from Fig. 2, $M(t)$ increases continuously as a function of time t. After waiting for one hour, $M(t)$ is still far from saturation. It has been commonly accepted that, if the long-time magnetic relaxation behavior occurs even in the applied magnetic fields much larger than the coercive field, the observed magnetic relaxation can be attributed to SG effect.[10] The best fit of the expression $M(t)=M_0+S\ln(t+t_0)$ to the experimental data yields the values of three fitting parameters, initial zero-field magnetization $M_0=0.5271$ emu/g, magnetic viscosity $S=0.0064$ emu/g, and characteristic time $t_0=91.8$ s.

The field cooling (FC) and zero-field cooling (ZFC) dc susceptibilities $\chi(=M/H)$ of U_2AuGa_3 were measured in various magnetic fields between 10 and 70 kOe. A part of the data is shown in Fig. 3. There exists a characteristic temperature $T_{\mathrm{ir}}(H)$, below which magnetic irreversibility manifesting as a bifurcation between the χ_{FC} and χ_{ZFC} curves is observed. The χ_{ZFC} curve in low applied field exhibits a sharp peak, which becomes broader, and its height decreases with increasing H. As typical features, these behaviors also signify the formation of SG state in U_2AuGa_3. The temperature $T_{\mathrm{ir}}(H)$ in 10 Oe is about 25.9 K, which varies as $-H^{2/5}$ and shifts to about 5.0 K in 70 kOe as clearly shown in the inset of Fig. 3. Note that the variation of T_{ir} with H does not follow the Almeida-Thouless Law (AT line: $T_{\mathrm{ir}}\propto -H^{2/3}$).[11] It should also be mentioned that, although the $H^{2/3}$-behavior of $T_{\mathrm{ir}}(H)$ was indeed observed for some uranium and rare earth SG materials, it was predicted for the case of an Ising-type SG in the infinite-range, random-bond mean-field model.[11] On the other hand, above 40 K, the χ_{ZFC} data in $H=100$ Oe (not shown here) can be nicely fitted using a modified Curie-Weiss Law with the effective magnetic moment $\mu_{\mathrm{eff}}=2.92\ \mu_{\mathrm{B}}$/U and the paramagnetic Curie temperature $\theta_p=14.7$ K. The obtained μ_{eff} value is much smaller than that ($\mu_{\mathrm{eff}}\sim 3.6\ \mu_{\mathrm{B}}$/U) expected for a free U ion with an f^2 or f^3 electronic configuration, indicating the itinerant behavior of $5f$ electrons and/or Kondo effect in this compound.

Our specific heat $C(T)$ (published in a conference proceeding[12] as the fragmentary data) and electrical resistivity $\rho(T)$ measurements are further evidence for the formation of the SG state in U_2AuGa_3. It is clear that both $C(T)$ (see Fig. 4) and $\rho(T)$ [see the inset (a) of Fig. 4] curves do not show any indication of a magnetic phase transition into a *long-range* order around T_f suggesting the absence of long-range spatial magnetic order in the vicinity of T_f. Moreover, at low temperatures the C/T vs T^2 plot [inset (b) of Fig. 4] yields for $T\rightarrow 0$ K a large γ value (the specific heat coefficient of T-linear term) of 86 mJ (mol U)$^{-1}$ K^{-2} as that usually observed in a common SG material. In addition, the

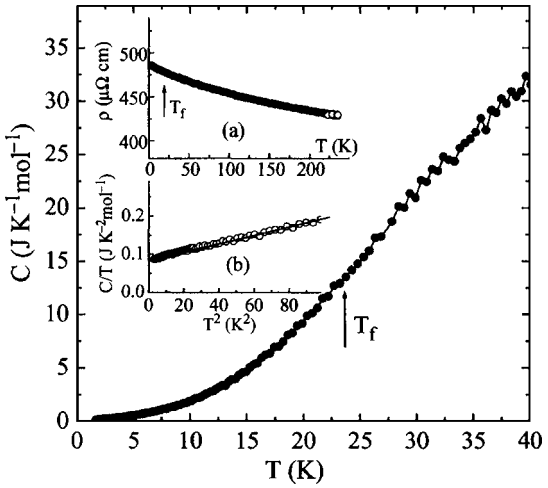

FIG. 4. Temperature dependence of specific heat of U_2AuGa_3 between 1.7 and 40 K. Inset (a): temperature dependence of electrical resistivity of U_2AuGa_3 between 4.2 and 240 K. Inset (b): specific heat data of U_2AuGa_3 plotted as C/T vs T^2.

negative slope in the $\rho(T)$ curve even at high temperatures is a common feature of uranium compounds. The large residual resistivity and the weak temperature dependence of $\rho(T)$ can be mainly ascribed to the structural disorder.

As described above, one necessary condition for a SG state is the existence of frustrated magnetic interactions. In contrast to the AlB_2-type SG materials such as U_2PdSi_3, in which U atoms form a triangular lattice and thus frustrated magnetic interactions can be induced due to the AF coupling between nearest neighbours,[1] U_2AuGa_3 crystallizes in the orthorhombic $CeCu_2$-type structure, and the triangular magnetic structure is not expected in this system. Thus how to understand the formation of frustrated magnetic interactions in such a system is an interesting problem. We point to a possible explanation as following. The disorder of nonmagnetic Au and Ga atoms in the crystal lattice of U_2AuGa_3 could destroy the long-range magnetic correlation and result in the formation of individual spins or finite-size granules with net magnetic moments (magnetic clusters). At low temperatures these randomly distributed clusters could interact with each other causing the formation of frustrated magnetic moments similar to what happen in amorphous or diluted metallic SG materials. In this context, we would like to compare the present results with those obtained for the isostructural compounds U_2PdGa_3 and U_2PtGa_3. The cluster model described above is also suitable for these two compounds. Compared with U_2AuGa_3, however, clusters with net magnetic moments in U_2PdGa_3 and U_2PtGa_3 seem to exist with much large geometric size probably due to the relatively strong magnetic interaction and/or different degree of randomness. Thus they show the signature of "almost long-range AF order" below T_N on the one hand, and lack discontinuity in both $C(T)$ and $\rho(T)$ curves at T_N with the addition of irreversible magnetic behavior at much lower temperatures originated from the "cluster effect" on the other hand.[3,4] In fact, based on NPD measurements, Tran *et al.*[4] estimated the AF correlation lengths in U_2PdGa_3 and U_2PtGa_3 to be 120–150 Å.

In conclusion, the $CeCu_2$-type uranium compound U_2AuGa_3 is confirmed to exist in the U_2TGa_3 family. This compound undergoes a SG transition with a static spin freezing temperature $T_s=23.6$ K. The up-shift of the ac susceptibility peak with increasing frequency, the down-shift of the dc susceptibility peak with increasing field, the long-time magnetic relaxation even in applied filed much large than the coercive field, the low temperature irreversible magnetism and the lack of any anomaly in both specific heat and electrical resistivity in the vicinity of T_s can be considered as the typical features characteristic of the SG state. We find that the variation of T_{ir} (the bifurcation point between the FC and ZFC susceptibilities) with H for U_2AuGa_3 is not consistent with the AT line, but follows a $T_{ir} \propto -H^{2/5}$ law. These behaviors could be understood in the framework of the magnetic cluster model. The spin freezing behavior found for U_2AuGa_3 in this work further richens the cooperative spin states discovered in the U_2TGa_3 family, that is to say varying from ferromagnetic to antiferromagnetic and to SG states with changing the transition metal.

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